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The use of scintillation counters in the measurement of the energy of X-rays and low energy gamma-rays

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With 10 figures in the text

Scintillation counters have of late become more widely used in physical research. One of the advantages is that they may be utilized for measuring the energy of gamma-rays. The pulse height distribution is determined and from this the different energies of the gamma-rays are found. Particularly valuable in this connection are phosphors containing atoms with a high atomic number, as, for instance, thallium-activated sodium iodide[1]. "Photo-electric peaks" are then obtained in the pulse height distribution which make possible a very satisfactory energy determination[2,3,4,5]. Earlier works in this field have almost entirely dealt with energies greater than 300–400 KeV. The aim of the work reported in this paper has been to study the possibility of measuring lower energies, down to 10 KeV, with the same method, as well as continuous X-ray spectra. The results obtained indicate that the scintillation counter is a useful instrument for the measurement of low energies.

Apparatus

Crystals of sodium iodide were prepared according to KYROPOULOS's[1] method, with a thallium content of 0.3 per cent in the melt. The crystals obtained were cleaved into smaller parts which were employed as phosphors. The crystal used in the present investigation was a 6.5 mm cube, absolutely clear. It was kept in paraffin oil. For the measurements it was taken out of the oil and was placed directly on the top of the photomultiplier. The oil that adhered to the crystal served as protection from the air and produced at the same time a good optical contact between the photomultiplier and the crystal. In order to collect the light emitted from the crystal the latter was surrounded by a cylindrical, thin-walled aluminum container, which was also placed directly on the top of the tube.

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[1] R. HOFSTADTER, Phys. Rev. *74*, 100 (1948).

[2] SVEN A. E. JOHANSSON, Nature *165*, 396 (1950); Arkiv för fysik *2*, 171 (1950).

[3] J. A. MCINTYRE & R. HOFSTADTER, Phys. Rev. *78*, 617 (1950).

[4] R. W. PRINGLE, K. I. ROULSTON & S. STANDIL, Phys. Rev. *78*, 627 (1950).

[5] P. R. BELL & I. M. CASSIDY, Phys. Rev. *79*, 173 (1950).

[1] S. KYROPOULOS, Z. Anorg. Chem. *154*, 308 (1926).

The photomultiplier used was of the type E.M.I. 5311. Owing to the fact that the photo-cathode is located immediately on the glass envelope a very good light collecting is obtained. The background is quite insignificant. Voltage to the dynodes was obtained from a voltage dividing network. The voltage was maintained at 130 volts per stage. The high voltage supply used[1] was very well stabilized. A change in the line voltage of 10 per cent altered the high voltage only 0.01 per cent. The stability towards changes in temperature was also very good.

The photomultiplier was enclosed in a thin-walled container of bakelite. The anode of the photomultiplier was connected to a cathode follower which was directly attached to the photomultiplier container.

The cathode follower was connected to an amplifier, the gain of which could be varied in steps. The amplifier was of the feed-back type and the plate voltage was well stabilized. In this manner one obtained good constancy in the gain.

The pulses from the amplifier were analyzed in a single channel differential discriminator of a type described by ELMORE and SANDS.[2] Studying the impulses with an oscilloscope proved to be a very convenient method for obtaining a rapid idea of the pulse distribution. For this purpose a Tektronix oscilloscope, type 511 A, was used. The pulse height was measured directly on the screen of the oscilloscope or through photographing the screen and subsequent measuring on the plates.

This apparatus was found to give measurements which were well reproducible. The values shown in the following curves were obtained on repeated occasions and the agreement was very good. The errors due to the statistical fluctuations are insignificant and they cannot influence in any way the shape of the curves obtained. They are therefore not drawn in the figures shown below.

Gamma-rays

Gamma-rays interact with matter through the photo-electric effect, the Compton effect, and pair production. Pair production occurs for energies greater than 1 MeV and is thus of no interest in this connection. In the photo-electric effect the photo-electron receives the energy of the photon minus the binding energy of the electron. When the electron shell is refilled following the emission of the photo-electron, X-rays are emitted. These X-rays can be wholly or partly absorbed by the crystal. When the crystal is large enough the entire energy of the photon is given off to it. The peak in the pulse height distribution corresponding to the photo-electric effect can therefore, under certain conditions, be assumed to correspond to the energy of the gamma-rays. In the Compton process, on the other hand, the emitted electrons have only a part of the energy of the gamma-rays, giving a continuous distribution in the pulse height distribution. In the Compton process the scattered rays can be absorbed within the crystal and the process thus contributes to the photo-electric peak, to an extent that is determined by the size of the crystal and the energy of the rays. With a sodium iodide crystal $3 \times 3 \times 3$ cm in size and energies below 300–400 KeV the scattered rays are almost entirely absorbed. With a crystal measuring $0.65 \times 0.65 \times 0.65$ cm, i.e., the size used in the present investigation, the limit lies around 150 KeV.

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[1] Designed by Dr. L. STIGMARK.

[2] W. C. ELMORE & M. SANDS, Electronics, Experimental Techniques (McGraw-Hill Book Company, Inc., 1949).

The best method for measuring the energy of gamma-rays is to determine the pulse height for the photo-electric peak. This is especially simple at low energies where the photo-electric effect dominates. The Compton distribution is very insignificant for energies below 150 KeV, and the scintillation counter should therefore be applicable in this energy region for measurements of the energy distribution in continuous X-rays. Examples of such measurements will be given below.

A question of great importance in this connection is that of resolution, which is determined by the spread in the pulse height. This spread is determined by several factors, from the light emission in the crystal to the amplification in the photomultiplier. It can be easily demonstrated, however, that the decisive factor is the statistical fluctuations in the number of electrons emitted from the photo-cathode of the multiplier. The fluctuations in the amplification of the multiplier also exert a certain influence. The peaks in the pulse distribution ought then to be Gaussian distributions, the width of which are largely determined by the number of electrons emitted from the photo-cathode. The resolution should thus be proportional to the square root of the mean of the electron number, i.e., proportional also to the square root of the energy. The curves presented below show that this is actually the case. The resolution can be determined from these curves and from this the number of electrons from the photo-cathode can be calculated. It appears then that an emitted energy of 100 KeV gives about 110 electrons. Naturally the figure varies with the crystal and photomultiplier employed. It agrees well with the value of 100 reported by HOFSTADTER and MCINTYRE.[1]

Before a scintillation counter can be used for energy measurements it must be calibrated through measuring with sources of known gamma-ray energies. It has previously been demonstrated[2] that the calibration curve is linear within the region of 0.4–3 MeV. In order to extend the calibration curve a few measurements were carried out with ^{131}J, whose gamma-ray energies are well known. The three energies 80, 163 and 364 KeV give three evenly distributed points for the drawing of a calibration curve. These points lie on a straight line passing through the origin. The calibration curve is thus linear down to the origin, i.e., the pulse height is proportional to the energy. This calibration curve was used in the energy determinations described on the following pages.

Measurements were carried out with the isotopes ^{141}Ce, ^{193}Os, and ^{203}Hg to enable a closer study of the behaviour of the "scintillation spectrometer" within the energy region 10–300 KeV. Small amounts of active material were fastened with zapon on a small piece of aluminum foil. The foil was thick enough to stop the beta-rays from the active material. This source was then mounted at a distance of 1 cm from the crystal. The rays were not collimated. Even very weak sources may be used with this method. The curves in the following were obtained with sources of 10^{-7} curie, or less. The time of measurement was one-half minute for each point. This source strength is no lower limit. Detailed curves can be obtained with considerably weaker sources, perhaps down to 10^{-9} curie. In such cases the time of measurement must be increased and greater statistical errors be permitted. The figures given are evident proof of the possibilities of the scintillation spectrometer for measuring extremely weak sources.

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[1] R. HOFSTADTER & J. A. MCINTYRE, Phys. Rev. *80*, 631 (1950).

[2] SVEN A. E. JOHANSSON, Nature *165*, 396 (1950).

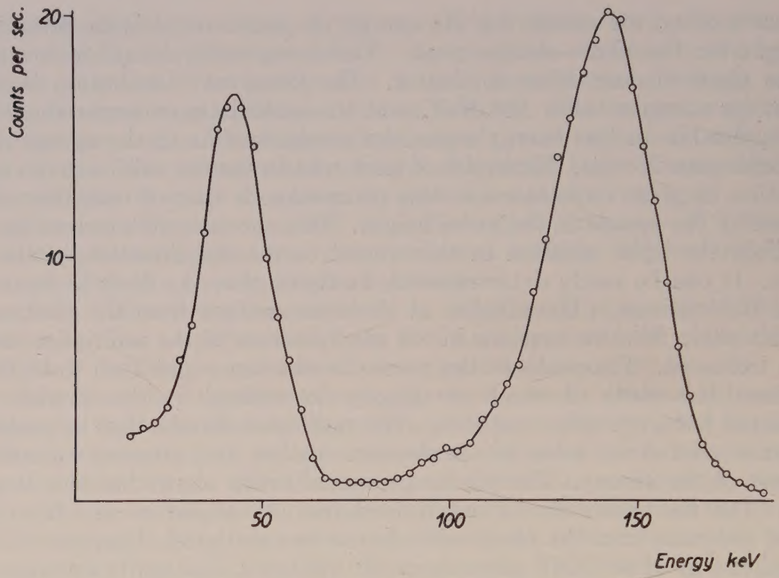

Fig. 1. Pulse height distribution for X-rays and gamma-rays of ^{141}Ce.

^{141}Ce

The radiation from this isotope has been recently studied by SHEPHERD[1], TER-POGOSSIAN et al.[2] and FREEDMAN and ENGELKEMEIR[3]. The energies of the gamma-rays were reported to be 141, 146, and 145 KeV, respectively. In addition, TER-POGOSSIAN reported a gamma-energy of 315 KeV. FREEDMAN and ENGELKEMEIR showed that in all probability this 315 KeV-radiation comes from ^{233}Pa, which results from neutron activation of thorium impurities in the cerium oxide.

^{141}Ce was obtained through neutron activation of cerium oxide for 4 weeks at Harwell. A small amount of the sample was mounted in the scintillation spectrometer as described above.

Figure 1 shows the pulse height distribution obtained by means of the differential discriminator. There is in the distribution a peak corresponding to an energy of 142 KeV, in good agreement with the values of the energy of the gamma-rays cited above. Since the gamma-rays are partly converted, the source also emits X-rays of praseodymium. The K X-rays correspond to the peak at 40 KeV. Due to the limited resolution no details in this peak can be observed. The measured value of 40 KeV deviates from the energy of the K_α X-rays of praseodymium which is 36 KeV. The deviation, however, probably lies within the limits of the experimental errors.

No gamma-rays could be detected at 315 KeV. At about 100 KeV, however, a very faint peak could be discerned, which was seen in all control measurements. It is not clear whether this 100 KeV-gamma radiation comes from ^{141}Ce or from

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[1] L. R. SHEPHERD, Research *1*, 671 (1948).

[2] TER-POGOSSIAN, COOK, GODDARD & ROBINSON, Phys. Rev. *76*, 909 (1949).

[3] M. S. FREEDMAN & D. W. ENGELKEMEIR, Phys. Rev. *79*, 897 (1950).

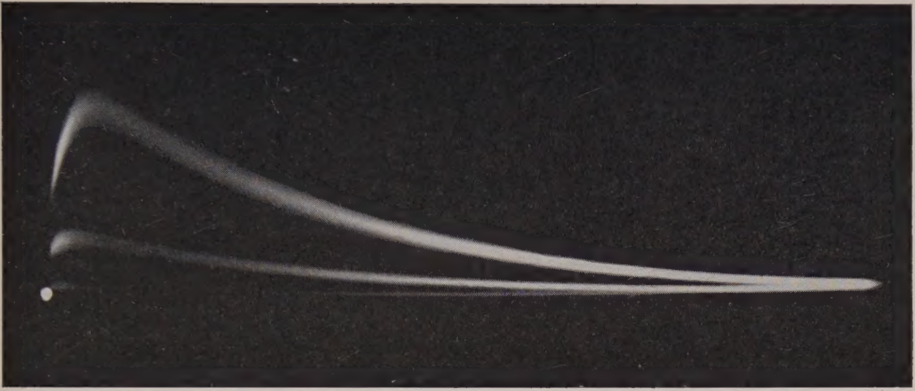

Fig. 2. The pulse height distribution for ^{141}Ce photographed on the oscilloscope screen.

some impurity. However, it decays with about the same half life as ^{141}Ce. On the curves over the photo-electron spectrum of ^{141}Ce, published by TER-POGOSSIAN et al.[1], two small peaks can be observed in this energy region.

The internal conversion coefficient for the K-shell can be computed from the pulse height distribution given in figure 1. The relation between the intensities of the K X-rays from praseodymium and the gamma-rays can be calculated from the curve. Here one must take into consideration the fact that the absorption of the sodium iodide crystal and the self-absorption of the source vary with the energy. It is also necessary to correct for the fluorescence yield. A conversion coefficient of 0.48 is then obtained. FREEDMAN and ENGELKEMEIR[2] report two values, 0.25 from spectrometer data and 0.7 from coincidence measurements.

Figure 2 shows a photograph of the oscilloscope screen with the pulses from ^{141}Ce. It appears from the picture that a very clear idea of the pulse distribution can be obtained with this simple method.

^{193}Os

When osmium is bombarded with slow neutrons three activities are obtained with the half-lives of 32 hours, 15 days, and 97 days. The main part of the gamma-rays is associated with the half-life of 15 days. These rays, which are attributed to the isotope ^{193}Os, have recently been studied by SAXON[3] and by BUNKER, CANADA, and MITCHELL[4]. Saxon reports the gamma-ray energies 127 and 39 KeV, Bunker et al. the energies 128 and 41 KeV.

Osmium powder was irradiated for 4 weeks at Harwell. A small part of the sample was examined in the scintillation spectrometer. Figure 3 shows the pulse height distribution obtained. The energy of the most energetic gamma-ray was found to be 124 KeV, in fairly good agreement with the measurements of the other investigators. As the gamma-ray is strongly converted, a marked peak is obtained cor-

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[1] TER-POGOSSIAN, COOK, GODDARD & ROBINSON, Phys. Rev. *76*, 909 (1949).

[2] M. S. FREEDMAN & D. W. ENGELKEMEIR, Phys. Rev. *79*, 897 (1950).

[3] D. SAXON, Phys. Rev. *74*, 264 (1948).

[4] M. E. BUNKER, R. CANADA & A. C. G. MITCHELL, Phys. Rev. *79*, 610 (1950).

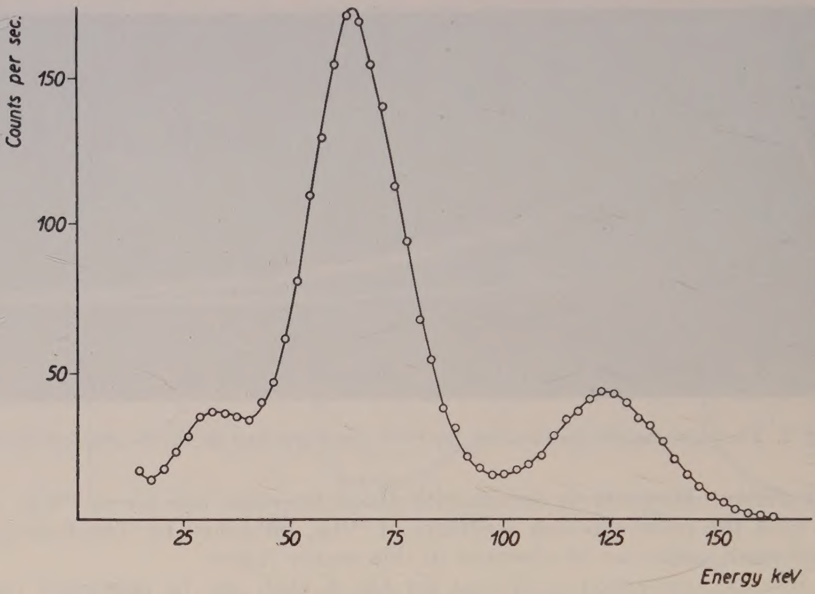

Fig. 3. Pulse height distribution for X-rays and gamma-rays of ^{193}Os.

responding to the K X-rays from iridium. The energy measured is 65 KeV, which is in exact agreement with the energy for the K_α X-rays of iridium.

The internal conversion coefficient for the K shell can also in this case be calculated from the pulse distribution. It should be kept in mind that the source also contains a small amount of the 97 day activity from ^{185}Os. The latter decays through K-capture, emitting in addition to gamma-rays also rhenium X-rays. The energy of the K_α X-rays from rhenium differs by only 3 KeV from the energy of the K_α X-rays of iridium and will thus contribute to the peak at 65 KeV. From the complete distribution curve, which also includes the gamma-rays from ^{185}Os at about 600 KeV, the relation between the two activities can be determined and corrections made for this effect. The internal conversion coefficient for the 124 KeV gamma-rays will then be 3.2.

At 35 KeV a minor peak is seen in the pulse distribution. Most likely it consists partly of Compton electrons from the 124 KeV gamma-rays. Here the Compton edge lies at 43 KeV. For the main part it must be the photo-electron peak from the 40 KeV gamma-rays. On account of this overlapping it is difficult to determine with any degree of accuracy the energy from the pulse distribution.

Figure 4 shows a photographic recording of the pulses on the oscilloscope screen. All of the details in the pulse distribution appear on this record.

^{203}Hg

On neutron irradiation of mercury activities result with the half-lives of 43 minutes, 23 hours, 64 hours, and 43 days. The 43 day activity has been attributed to

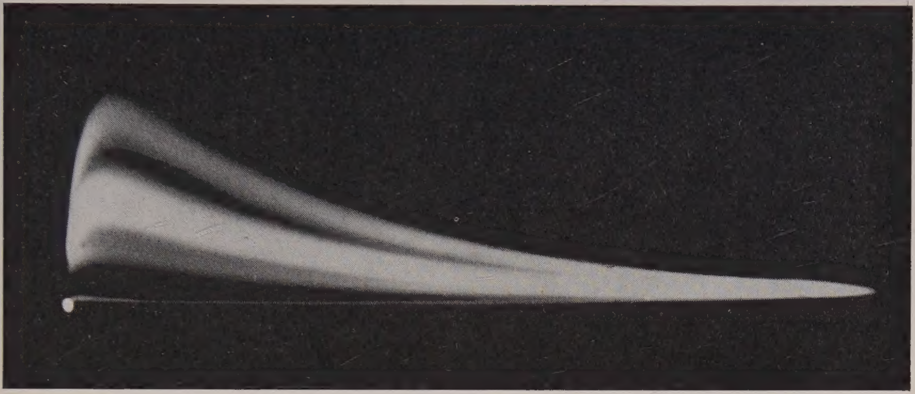

Fig. 4. The pulse height distribution for ^{193}Os photographed on the oscilloscope screen.

^{203}Hg by SLÄTIS and SIEGBAHN.[1] The radiation from this isotope has been recently studied by SAXON[2] and by SLÄTIS and SIEGBAHN[1]. It decays through a simple beta-transition followed by a single gamma-ray. Saxon reports the energy of the gamma-ray to be 286 KeV, SLÄTIS and SIEGBAHN 279 KeV.

Mercury oxide was irradiated at Harwell for 4 weeks. After the shorter activities had decayed a small amount of the sample was examined in the scintillation spectrometer. The pulse distribution curve appears in figure 5. The photo-electron peak of the gamma-rays gives the energy of 286 KeV. The peak at 75 KeV is attri-

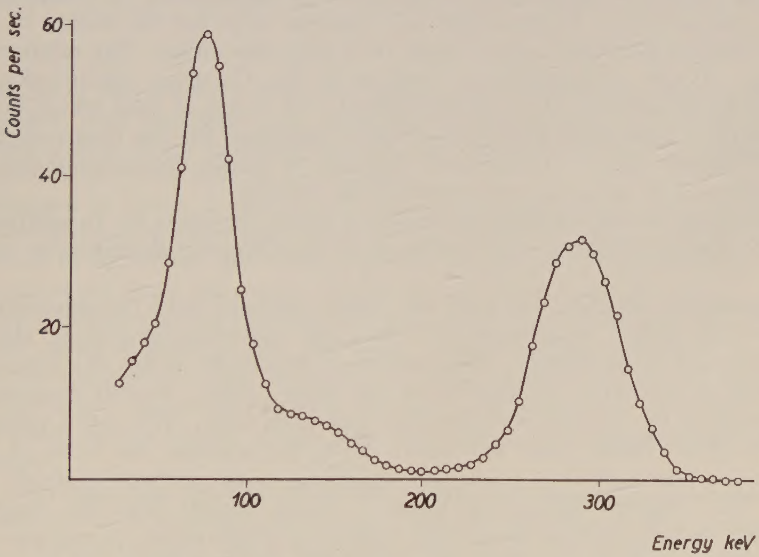

Fig. 5. Pulse height distribution for X-rays and gamma-rays of ^{203}Hg.

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[1] H. SLÄTIS & K. SIEGBAHN, Phys. Rev. *75*, 318 (1949).

[2] D. SAXON, Phys. Rev. *74*, 849 (1948).

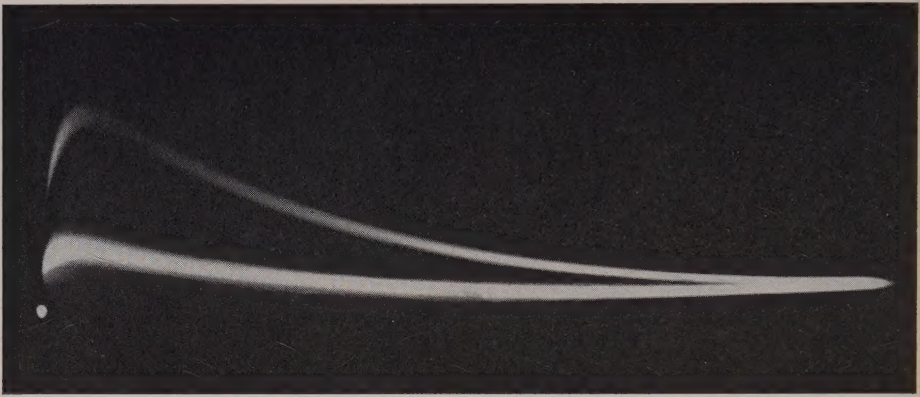

Fig. 6. The pulse height distribution for ^{203}Hg photographed on the oscilloscope screen.

buted to the X-rays following the internal conversion of the gamma-rays. The K_α X-rays from thallium have the energy of 73 KeV. From the pulse distribution curve the internal conversion coefficient for the K-shell can be calculated. For this purpose it is necessary to know the absorption in the crystal of the gamma-rays; the X-rays can be assumed to be absorbed completely. On the basis of the absorption coefficient for lead[1] it was computed that 23 per cent of the 286 KeV gamma-rays yield photo-electrons in the sodium iodide crystal used. This gives an internal conversion coefficient of 0.21. The value given by Saxon is 0.18.

It appears from the pulse distribution curve that the X-ray peak lies over a continuous distribution which is interpreted as a distribution of Compton electron from the gamma-rays. Theoretically the Compton edge has the energy of 150 KeV. The distribution measured agrees fairly well with this value. The relation between the areas of the photo-electron peak and the Compton distribution can be estimated from figure 5. It is approximately the same as that which can be calculated with a knowledge of the absorption coefficient for the photo-electric effect and the Compton effect. As has been pointed out above, this relationship is somewhat influenced by secondary processes in the crystal.

A photograph of the oscilloscope screen is shown in figure 6. In addition to the two photo-electric peaks, a faint streak from the Compton distribution can be discerned.

The agreement obtained between the values obtained with the scintillation spectrometer and earlier measurements with beta spectrometers show the calibration curve used to be correct. This is further proved by means of a diagram where the energy is plotted as a function of the pulse height. In this diagram, energy values reported by various investigators have been used. When the values do not agree a weighted mean value was used. Along the abscissa the value of the pulse height of the photo-electron peak, corresponding to the various energies, has been plotted. The diagram appears in figure 7. It is evident from the diagram that the relation between pulse height and energy is linear down to energies of a few kilovolts. It should be observed, however, that the appearance of the calibration curve depends on the voltage on the multiplier. Too high voltage produces an in-

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[1] L. G. Glendenin, Nucleonics *2*, 1 (1948).

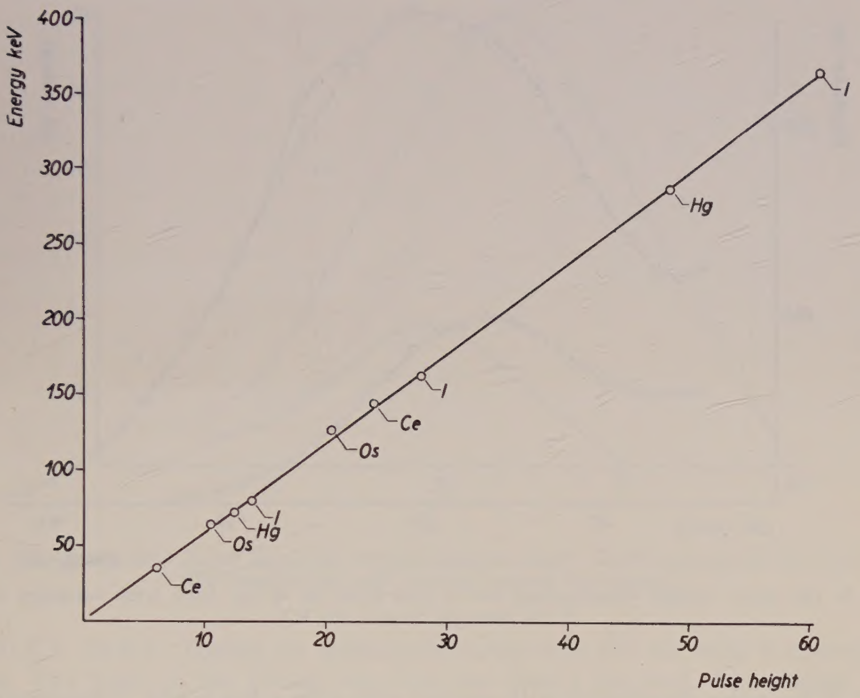

Fig. 7. The energy of X-rays and gamma-rays of ^{131}I, ^{141}Ce, ^{193}Os and ^{203}Hg plotted against the pulse height of the photo-electric peaks.

creased slope of the curve so that the pulse height becomes proportionally less for high energies. This change is not caused by the amplifier or the discriminator but by the multiplier. Some multipliers were also observed to give an increased slope of the calibration curve even at low voltages.

The diagram in figure 7 also gives an idea of the accuracy of the method. The average deviation from the straight line is about 2 per cent. If the energy values used are assumed to be correct this figure provides a measure of the accuracy. This can also be estimated from the pulse height distributions reported above. It should be possible to determine the position of a peak with an error of less than 2 per cent, at least for energies greater than 100 KeV. The remaining part of the total error is caused by changes in the electronic circuits, particularly in the zero-point of the discriminator. It seems possible to make improvements on these points, so as to increase the accuracy above that attained in the measurements published in this paper.

When comparing the scintillation spectrometer with magnetic spectrometers, the former has evident advantages, primarily its great sensitivity. The sources can be mounted in such a manner that a very large part (up to 50 per cent) of the radiation strikes the crystal. In the energy region studied here, up to 100 per cent of the radiation is absorbed even by a comparatively small crystal. It follows that even very weak sources can be measured. There is no difficulty in obtaining a good pic-

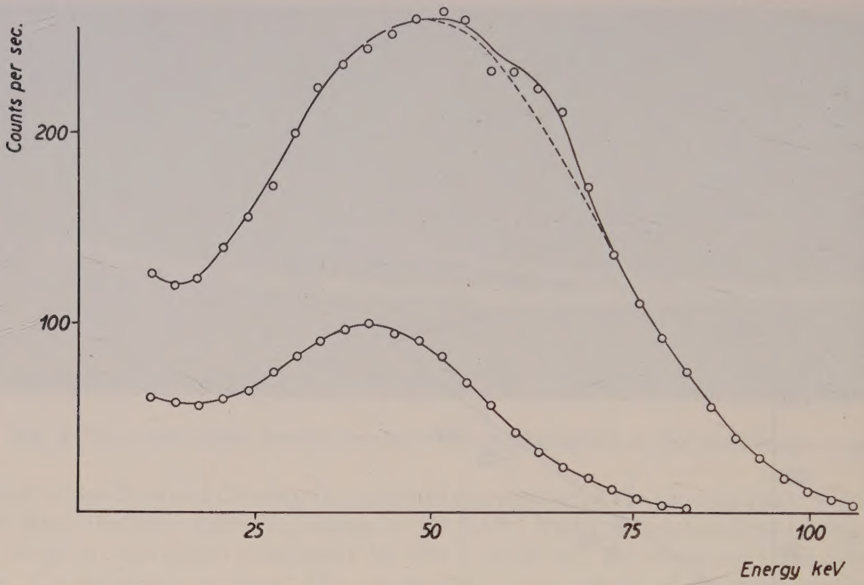

Fig. 8. The pulse height distributions for X-rays from an X-ray tube with tungsten target. Tube voltages 75 and 100 KV.

ture of the pulse distribution with sources with only a few tenths of disintegrations per second, i.e., the strength of 10^{-9} curie.

Another advantage is the possibility of measuring also very low energies, down to about 10 KeV.

The greatest drawback of the method is its low resolution. It is, however, probably sufficient for most gamma spectra.

Finally it should be stressed that the apparatus here described is very simple. Nearly all the components are commercially available. It is strongly constructed and functions reliably.

X-rays

The scintillation spectrometer should also be utilizable for the measurement of continuous X-ray spectra. In this case it is necessary to know the relation between pulse height and energy as well as the pulse height distribution for monoenergetic radiation. The measurements in the preceding section give the information desired. The pulse height is directly proportional to the energy. Monoenergetic radiation gives a Gaussian distribution. The width of the distribution can be determined from these measurements. There is no continuous Compton distribution below 125 KeV.

In order to test this possibility in practice the pulse height distribution from an ordinary X-ray tube was measured at different voltages. The tube used was of the type Philips Metwa Metallix, 220 KV, 12 mA, with a tungsten target. The highvoltage supply employed was of the Greinacher type with a maximal effect

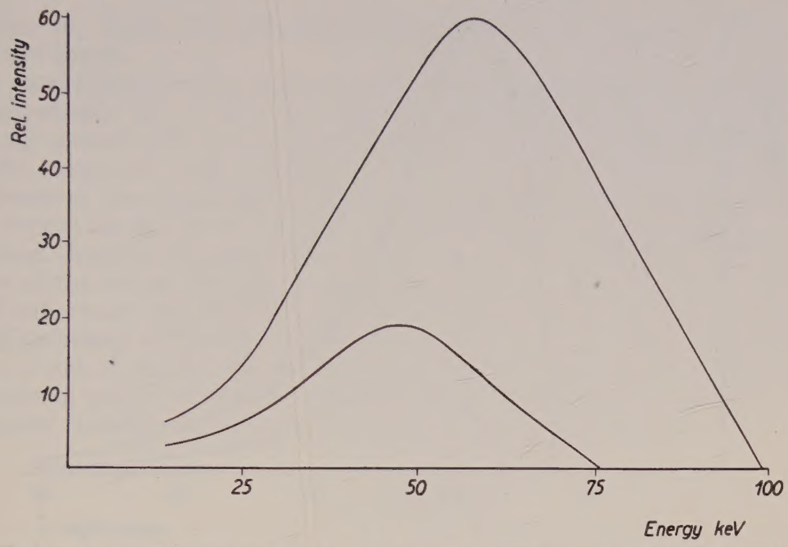

Fig. 9. The continuous X-ray spectrum from a tungsten target. Tube voltages 75 and 100 KV. The relative intensity plotted against the energy.

of 200 KV, 30 mA. During the measurements the tube was run with a current of 4 mA. The ripple in the voltage was then less than 2 per cent. The voltage was measured with a voltmeter across the primary terminals of the transformer. The voltmeter was calibrated with a spark gap. The scintillation spectrometer was placed at a distance of 5 meters from the X-ray tube. The radiation was collimated by means of three lead plates, the last of which had a circular hole with a diameter of 0.1 mm. It was placed at a distance of 1 cm from the crystal.

The X-rays measured were filtered considerably, both in the target of the tube and in the tube window, which had an aluminum equivalent of 1.5 mm. The purpose of the measurement was, however, to determine the energy distribution of the rays emitted by the tube. No corrections were therefore attempted for this absorption.

Figure 8 shows the pulse distribution curves for the two voltages 75 and 100 KV. The curve for 75 KV has an even course but the curve for 100 KV becomes irregular approximately at the energy of 60 KeV. This irregularity lies entirely outside the statistical fluctuations. In other measurements, moreover, with different voltages up to 150 KeV an irregularity has always been obtained at 60 KeV. This effect is interpreted as an overlapping on the continuous spectrum of a peak from the K X-rays from tungsten. Due to the low resolution no details can be seen in the K X-rays. At the voltage of 75 KV, the intensity of the K X-rays is very insignificant and causes no peak in the pulse distribution. The broken line in the 100 KV-curve shows the shape assumed for the continuous X-ray spectrum within this energy region. This extrapolation naturally involves some uncertainty but it should not be too great.

When a continuous distribution is measured the curves obtained are affected by the resolution of the apparatus. This effect is negligible only in apparatuses of very great resolution. For a scintillation spectrometer with its limited resolution

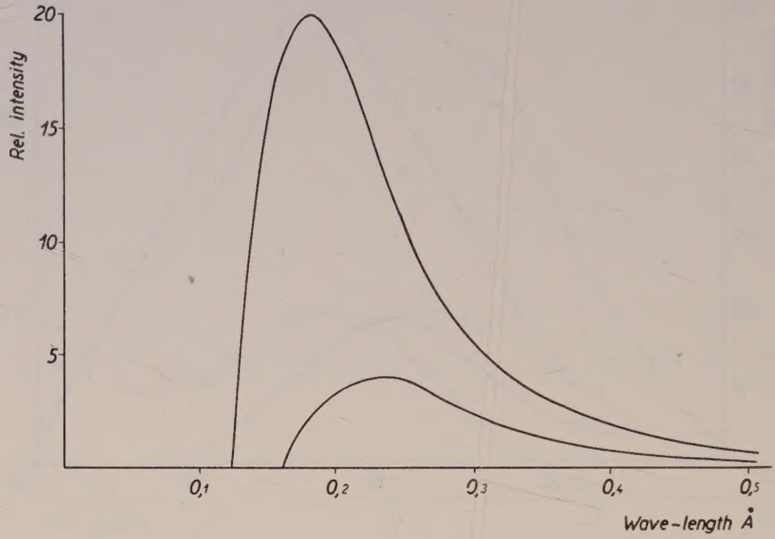

Fig. 10. The curves in fig. 9 with the relative intensity plotted against the wave-length.

the effect is important, as appears from the upper limit for the curves in figure 8. The pulse distributions extend beyond 75 and 100 KeV, respectively, approaching the axis asymptotically. A correction must therefore be made for this effect. OWEN and PRIMAKOFF[1] have treated this case for magnetic spectrometers. By means of suitable approximations they arrived at a simple expression of the correction term. At low resolution, however, the approximations become too inaccurate. The curves in figure 8 have instead been corrected by means of graphic integration. The pulse distribution for monoenergetic radiation at different energies is known from the measurements described in the foregoing section. It is then possible to determine graphically the deformation of a given pulse distribution caused by the limited resolution. This deformation, expressed in per cent, is fairly insignificant, except for the uppermost part of the curve. The correction can therefore be made with fairly great accuracy.

The corrected curves are seen in figure 9. They are given in the form of intensity as a function of the energy. It is a well-known fact that in such a representation the continuous X-ray spectrum is linear in its uppermost part. The curves given in figure 9 have exactly this shape which enables a fairly accurate determination of the energy limit and thus also of the voltage of the X-ray tube. The curves do not cut the axis at exactly 75 and 100 KeV, partly on account of the inaccuracy of the apparatus itself and partly because the voltage of the X-ray tube could not be set and maintained constant with any greater degree of precision.

The absorption of the X-rays in target and tube-window causes the curves to deflect from the straight lines at lower energies. The absorption has also the effect that the total intensity at 75 KV is only one-fourth of that at 100 KV. Without the absorption one would have expected the relation to be approximately 1 : 2.

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[1] G. E. OWEN & H. PRIMAKOFF, Phys. Rev. *74*, 1406 (1948).

In figure 10, finally, the two curves are given with the intensity as a function of the wave-length.

These measurements show that, in spite of the limited resolution, the scintillation spectrometer can be used for studying continuous X-ray spectra. As in the case of the gamma-rays, the greatest advantage of the spectrometer is its great sensitivity, even very weak intensities can be measured, as, for example, scattered rays. Moreover, the sensitivity is constant (100 %) up to at least 125 KeV. With a larger crystal one can go even higher. An upper limit is attained when the number of Compton electrons becomes equal to the number of photo-electrons. An interpretation of the curves obtained is then very difficult. This limit lies at 200 to 250 KeV. Furthermore the sensitive volume is small. Details can be studied in the spatial distribution of X-rays. When the crystal is mounted on a light guide, for instance, a rod of quartz, the intensity and the energy distribution can be determined even in places which are difficult to access. The method described in this paper should therefore prove quite useful in the field of radiology.

Acknowledgement

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Department of Physics, University of Lund, February 1951.

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Uppsala 1951. Almqvist & Wiksells Boktryckeri AB